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(54) FLAME-RETARDANT POLYCARBONATE RESIN COMPOSITION

(57)Abstract:

PROBLEM TO BE SOLVED: To obtain a flame-retardant polycarbonate resin composition having excellent heat resistance and water vapor resistance which excels in mechanical properties, flame retardance, impact resistance, and flowability and can maintain high physical properties even at high temperatures and high humidities.

SOLUTION: This flame-retardant polycarbonate resin composition comprises (a) 100 pts.wt. aromatic polycarbonate resin, (b) 5-100 pts.wt. aromatic vinyl/vinyl cyanide based copolymer, (c) 1-10 pts.wt. multi-layer structure polymer, (d) 0.5-40 pts.wt. phosphazene compound, and (e) 0.01-5 pts.wt. polyfluoroethylene.

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CLAIMS

[Claim(s)]

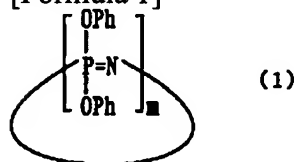
[Claim 1] (a) The fire-resistant polycarbonate resin constituent which comes to blend (b) aromatic series vinyl / vinylcyanide system copolymer 5 - the 100 weight sections, (c) multilayer-structure polymer 1 - 10 weight sections, (d) phosphazene compound 0.5 - 40 weight sections, and (e) poly fluoro ethylene 0.01 - 5 weight sections with the aromatic series polycarbonate resin 100 weight section.

[Claim 2] (b) The fire-resistant polycarbonate resin constituent according to claim 1 whose aromatic series vinyl / vinylcyanide system copolymer of a component are graft copolymers which come to carry out the polymerization of the bottom styrene monomer of existence of rubber, and the acrylonitrile (meta) system monomer.

[Claim 3] (b) The fire-resistant polycarbonate resin constituent according to claim 1 whose aromatic series vinyl / vinylcyanide system copolymer of a component are mixture with the copolymer of the graft copolymer which comes to carry out the polymerization of the bottom styrene monomer of existence of rubber, and (meta) the acrylonitrile system monomer, a styrene monomer, and (meta) an acrylonitrile system monomer.

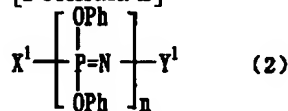
[Claim 4] (c) The phosphazene compound of a component is a general formula (1).

[Formula 1]



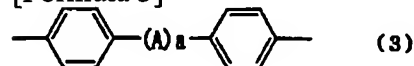
The integer of 3-25 is shown by the inside m of [type. Ph shows a phenyl group.] The annular phenoxy phosphazene, the general formula (2) which are come out of and expressed

[Formula 2]



Radical-N=P (OPh)3 or radical-N=P(O) OPh is shown by the inside X1 of [type, and Y1 is a radical. - P(OPh) 4 or radical-P(O) (OPh)2 are shown. n shows the integer of 3-10000. Ph is the same as the above.] At least one sort of phosphazene compounds chosen from the group which comes out and becomes the chain-like phenoxy phosphazene expressed and a list from said annular phenoxy phosphazene and said chain-like phenoxy phosphazene are o-phenylene group, m-phenylene group, p-phenylene group, and a general formula (3).

[Formula 3]



-C(CH3)2-, -SO2-, -S-, or -O- is shown by the inside A of [type. a shows 0 or 1.] It is the compound over which at least one sort of bridge formation radicals chosen from the group which comes out and consists of a bis-phenylene group expressed come to construct a bridge. (i) This bridge formation radical intervenes between two oxygen atoms from which the phenyl group of a phosphazene compound was desorbed. (ii) The content percentage of a phenyl group

is 50 - 99.9% on the basis of the total of the above-mentioned phosphazene compound (1) and/or all the phenyl groups in (2). And (iii) the fire-resistant polycarbonate resin constituent according to claim 1 characterized by being at least one sort chosen as intramolecular from the group of a bridge formation phenoxy phosphazene compound which does not have the free hydroxyl group.

[Claim 5] (c) Claim 1 in which a multilayer-structure polymer contains an alkyl (meta) acrylate system polymer, claim 2, a fire-resistant polycarbonate resin constituent according to claim 3 or 4.

[Claim 6] (c) The fire-resistant polycarbonate resin constituent according to claim 5 with which a multilayer-structure polymer contains a diene system polymer.

[Claim 7] (a) Claim 1 characterized by the viscosity average molecular weight of the aromatic series polycarbonate resin of a component being 16000-30000, claim 2, a fire-resistant polycarbonate resin constituent according to claim 3 or 4.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] In more detail, fabrication nature is good and this invention relates to a fire-resistant polycarbonate resin constituent and the improved heat-resistant, shock-proof, and damp-proof fire-resistant polycarbonate resin constituent.

[0002]

[Description of the Prior Art] Conventionally, PC resin constituent which blended styrene / acrylonitrile system copolymers, such as ABS plastics, with polycarbonate resin ("PC resin" may be called below) is used in the extensive field. It is such a resin constituent and the ingredient with which the phosphorus series flame retardant was blended with the resin constituent of PC resin and ABS plastics is indicated by JP,2-115262,A, JP,2-32154,A, etc. as a fire-resistant ingredient which can be used for the application of OA equipment etc.

[0003] However, these flame retardant materials were insufficient in moisture resistance. So, with the technique given in JP,11-130954,A, the moisture resistance of a resin constituent is improved by an alkali-metal salt content using ABS 1 ppm or less, and using phosphoric ester with the low acid number. Furthermore, the constituent which improved the moisture resistance of the constituent containing phosphoric ester is indicated by JP,11-189714,A by adding the acid scavenger represented by epoxy, oxazoline, and ortho ester.

[0004] However, in these constituents, since it is not what improves the hydrolysis nature of the flame retarder resulting from the structure of the flame retarder to be used itself, it has not resulted until it improves the moisture resistance of this constituent sharply.

[0005] Furthermore, in the fire-resistant constituent of a PC/ABS alloy, in order to secure a fluidity, the molecular weight of PC resin may be lowered and it becomes difficult only with the conventional ABS plastics to secure shock resistance in that case.

[0006]

[Problem(s) to be Solved by the Invention] This invention makes it a technical problem to offer fire-resistant PC resin constituent excellent in thermal resistance or moisture resistance which is excellent in fire retardancy, shock resistance, and a fluidity, and can maintain high physical properties an elevated temperature and under highly humid.

[0007]

[Means for Solving the Problem] In order that this invention person may solve the above-mentioned technical problem, as a result of repeating research wholeheartedly, in the (a) aromatic series polycarbonate resin 100 weight section (b) Aromatic series vinyl / vinylcyanide system copolymer 5 - the 100 weight sections, (c) It found out that the fire-resistant polycarbonate resin constituent which comes to blend the multilayer-structure polymer 1 - 10 weight sections, (d) phosphazene compound 0.5 - 40 weight sections, and (e) poly fluoro ethylene 0.01 - 5 weight sections could turn into a desired fire-resistant polycarbonate resin constituent. This invention is completed based on this knowledge.

[0008] That is, this invention relates to the fire-resistant polycarbonate resin constituent which comes to blend (b) aromatic series vinyl / vinylcyanide system copolymer 5 - the 100 weight sections, (c) multilayer-structure polymer 1 - 10 weight sections, (d) phosphazene compound 0.5 - 40 weight sections, and (e) poly fluoro ethylene 0.01 - 5 weight sections with the (a) aromatic series polycarbonate resin 100 weight section.

[0009]

[Embodiment of the Invention] The thermoplastic aromatic series polycarbonate polymer or copolymer which is made

as (a) aromatic series polycarbonate resin in this invention by for example making an aromatic series hydroxy compound, or this and a little polyhydroxy compound react with the diester of a phosgene or carbonic acid and which may branch can be mentioned. About the manufacture approach of this polycarbonate polymer or a copolymer, it is not limited and can manufacture by the various approaches that the phosgene method (interfacial polymerization), scorification (ester interchange method), etc. are conventionally well-known.

[0010] Moreover, in this invention, the aromatic series polycarbonate resin which was manufactured with scorification and which adjusted the amount of OH radicals of an end group can be used.

[0011] As an aromatic series dihydroxy compound, 2 and 2-screw (4-hydroxyphenyl) propane (= bisphenol A), tetramethyl bisphenol A, screw (4-hydroxyphenyl)-p-diisopropylbenzene, hydroquinone, resorcinol, 4, and 4-dihydroxydiphenyl etc. is mentioned, and bisphenol A is mentioned preferably.

[0012] In order to obtain branched aromatic series polycarbonate resin Phloroglucine, 4, 6-dimethyl - 2, 4, 6-Tori (4-hydroxyphenyl) heptene - 2, 4, 6-dimethyl - 2, 4, 6-Tori (4-hydroxyphenyl) heptane, 2 and 6-dimethyl - 2, 4, and 6-Tori (4-hydroxyphenyl) heptene -3, 1 and 3, and 5-Tori (4-hydroxyphenyl) benzene --) Polyhydroxy compounds, such as 1, 1, and 1-Tori (4-hydroxyphenyl) ethane, Or what is necessary is just to follow the above-mentioned approach, using 3 and 3-screw (4-hydroxy aryl) oxy-indole (= isatin bisphenol), 5-KURORU isatin, 5, a 7-dichloro isatin, 5-bromine isatin, etc. as said some of aromatic series dihydroxy compounds. the amount of these compounds used -- an aromatic series dihydroxy compound -- receiving -- usually -- 0.01-10-mol % -- it is 0.1-2-mol % preferably.

[0013] In order to adjust the molecular weight of aromatic series polycarbonate resin, m- and p-methyl phenol, m- and p-propyl phenol, p-tert-butylphenol, a p-long-chain alkylation phenol, etc. are mentioned that what is necessary is just to use a monovalence aromatic series hydroxy compound.

[0014] The polycarbonate copolymer guided as desirable aromatic series polycarbonate resin from polycarbonate resin [which is guided from 2 and 2-screw (4-hydroxyphenyl) propane] or 2, and 2-screw (4-hydroxyphenyl) propane and other aromatic series dihydroxy compounds, for example is mentioned.

[0015] In this invention, these aromatic series polycarbonate resin is one-sort independent, or two or more sorts can use it, mixing.

[0016] The molecular weight of aromatic series polycarbonate resin is the viscosity average molecular weight converted from the solution viscosity measured at the temperature of 25 degrees C, using a methylene chloride as a solvent, is 16000-30000 preferably, and is 18000-23000 more preferably.

[0017] As the aromatic series vinyl / a vinylcyanide system copolymer of the (b) component in this invention, the graft copolymer which comes to carry out the graft polymerization of the copolymer of a styrene monomer and (meta) an acrylonitrile system monomer, the bottom styrene monomer of existence of rubber, and (meta) the acrylonitrile system monomer, for example is mentioned.

[0018] As a copolymer of a styrene monomer and an acrylonitrile (meta) system monomer, an AS resin etc. is mentioned, for example. Moreover, as a graft copolymer which carried out the graft polymerization of the bottom styrene monomer of existence of rubber, and the acrylonitrile (meta) system monomer, ABS plastics, AES resin, AAS resin, etc. are mentioned, for example. Generally as the manufacture approach of a styrene system copolymer, various kinds of approaches, such as an emulsion-polymerization method, a solution polymerization method, a suspension-polymerization method, and a bulk-polymerization method, are learned. or [that there are few amounts of survival of an alkali-metal salt (used as the emulsifier in an emulsion-polymerization method, a dispersant in a suspension-polymerization method, etc.) as a copolymer of the styrene monomer and acrylonitrile (meta) system monomer which are used for this invention] -- or it is desirable to use what is not included. It is desirable to use the aromatic series vinyl / vinylcyanide system copolymer manufactured by the bulk-polymerization method in this invention from this viewpoint.

[0019] As the aromatic series vinyl / a vinylcyanide system copolymer in this invention, mixture with the copolymer of the graft copolymer which comes to carry out the graft polymerization of the bottom styrene monomer of existence of rubber and (meta) the acrylonitrile system monomer, the graft copolymer which comes to carry out the polymerization of the bottom styrene monomer of existence of rubber and (meta) the acrylonitrile system monomer to a list, a styrene monomer, and (meta) an acrylonitrile system monomer etc. is mentioned preferably.

[0020] In the above-mentioned graft copolymer or the copolymer of a styrene monomer and (meta) an acrylonitrile system monomer, the monomer in which these and copolymerization are possible may contain as a comonomer with the styrene monomer and the acrylonitrile (meta) system monomer.

[0021] Here, as a styrene monomer, styrene, alpha methyl styrene, p-methyl styrene, etc. are mentioned, and styrene is mentioned preferably, for example. As a styrene monomer, an acrylonitrile (meta) system monomer, and a monomer that can be copolymerized, acrylic-acid (meta) alkyl ester, such as a methyl acrylate, an ethyl acrylate, acrylic-acid propyl, a methyl methacrylate, and ethyl methacrylate, maleimide, N-phenyl maleimide, etc. are mentioned, and acrylic-acid (meta) alkyl ester is mentioned preferably, for example.

[0022] As rubber, rubber 10 degrees C or less is preferably mentioned for glass transition temperature. As an example of this rubber, diene system rubber, acrylic rubber, ethylene/propylene rubber, silicone rubber, etc. are mentioned, for example, and diene system rubber, acrylic rubber, etc. are mentioned preferably.

[0023] As diene system rubber, polybutadiene, a butadiene / styrene copolymer, polyisoprene, the low-grade alkyl ester copolymer of a butadiene/(meta) acrylic acid, the low-grade alkyl ester copolymer of a butadiene / styrene / (meta) acrylic acid, etc. are mentioned, for example. Here, as low-grade alkyl ester of an acrylic acid (meta), a methyl acrylate, an ethyl acrylate, a methyl methacrylate, ethyl methacrylate, etc. are mentioned, for example. As for the rate of the low-grade alkyl ester of the acrylic acid in the low-grade alkyl ester copolymer of a butadiene/(meta) acrylic acid, or the low-grade alkyl ester copolymer (meta) of a butadiene / styrene / (meta) acrylic acid, it is desirable that it is 30 or less % of the weight of rubber weight.

[0024] As acrylic rubber, acrylic-acid alkyl rubber is mentioned and the carbon numbers of an alkyl group are 1-8 preferably, for example. As an example of acrylic-acid alkyl rubber, an ethyl acrylate, butyl acrylate, acrylic-acid ethylhexyl, etc. are mentioned. The ethylenic unsaturated monomer of cross-linking may be used for arbitration, and alkylene diol, di(meth)acrylate, polyester TERUJI (meta) acrylate, a divinylbenzene, trivinylbenzene, a cyanuric acid triaryl, an acrylic-acid (meta) allyl compound, a butadiene, an isoprene, etc. are mentioned to acrylic-acid alkyl rubber as a cross linking agent, for example. As acrylic rubber, the core shell mold polymer which has bridge formation diene system rubber as a core is mentioned further.

[0025] The loadings of the aromatic series vinyl / vinylcyanide system copolymer in this invention are the 5 - 100 weight section to the aromatic series polycarbonate resin 100 weight section. A fluidity tends to fall that the loadings of aromatic series vinyl / vinylcyanide system copolymer are under 5 weight sections, and if the 100 weight sections are exceeded, thermal resistance will tend to fall. The loadings of aromatic series vinyl / vinylcyanide system copolymer are 6 - 70 weight section preferably to the aromatic series polycarbonate resin 100 weight section, and are 7 - 50 weight section still more preferably.

[0026] The multilayer-structure polymer of the (c) component in this invention means the polymer with which it comes to carry out sequential covering of the polymer which constitutes an innermost nuclear lamina (heart) in 1 or two or more outer core layers. As this multilayer-structure polymer, the multilayer-structure polymer containing an alkyl (meta) acrylate system polymer etc. is mentioned, for example. Furthermore, the multilayer-structure polymer which contains diene system polymers, such as polybutadiene, and styrene / butadiene copolymer, with an alkyl (meta) acrylate system polymer is desirable.

[0027] These multilayer-structure polymers can be manufactured by multistage story seed polymerization to which the polymer of a next phase carries out sequential covering of the polymer of a previous phase and which continued.

[0028] As a component which forms the innermost nuclear lamina of these multilayer-structure polymer, glass transition temperature can illustrate a bridge formation component 0 degree C or less preferably. As a bridge formation component, these are independent one sort, or bridge formation rubber components, such as a bridge formation rubber component of a butadiene, and styrene/butadiene, the bridge formation rubber component of an alkyl (meta) acrylate system polymer, polyorganosiloxane, etc. can be mentioned, for example, and they are used by two or more sorts here, mixing.

[0029] Moreover, as a component which forms the outermost nuclear lamina, an aromatic series vinyl monomer, non-aromatic series system monomers, or two or more kinds of such mixture are mentioned. As an aromatic series vinyl monomer, styrene, vinyltoluene, alpha methyl styrene, mono-KURORU styrene, dichloro styrene, bromostyrene, etc. are mentioned, for example. Also in these, especially styrene is used preferably. As a non-aromatic series system monomer, vinylcyanides, cyanidation vinylidenes, etc., such as alkyl methacrylate, such as alkyl acrylate, such as ethyl acrylate, butyl acrylate, and ethylhexyl acrylate, methyl methacrylate, and butyl methacrylate, acrylonitrile, and a methacrylonitrile, are mentioned, for example. As for the carbon number of the alkyl group in alkyl acrylate or alkyl methacrylate, one to about eight are good.

[0030] In manufacturing an acrylic-acid alkyl polymer or an alkyl methacrylate polymer using alkyl acrylate or alkyl

methacrylate, cross linking agents, such as an ethylenic unsaturated monomer, may be used. As this cross linking agent, alkylene diol, di(meth)acrylate, polyester TERUJI (meta) acrylate, a divinylbenzene, trivinylbenzene, a cyanuric acid triaryl, an acrylic-acid (meta) allyl compound, etc. are mentioned, for example.

[0031] In the multilayer-structure polymer which has one or more interlayers between an innermost nuclear lamina and the outermost nuclear lamina, the polymer which constitutes an aforementioned innermost nuclear lamina or the aforementioned outermost nuclear lamina can be suitably used as a polymer which constitutes an interlayer.

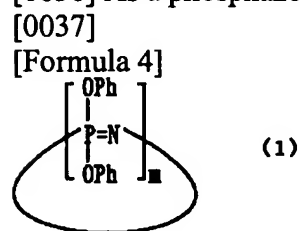
[0032] Furthermore, a constituent without a poor appearance (pearl gloss etc.) can be obtained by using the multilayer-structure polymer currently indicated by JP,5-222140,A.

[0033] As a multilayer-structure polymer of this invention, poly alkyl (meta) acrylate which is indicated by JP,5-339462,A can be made into an innermost nuclear lamina, and the multilayer-structure polymer which has the second layer which consists of the first pass and the poly alkyl (meta) acrylate which consist of polyorganosiloxane on it can also be used.

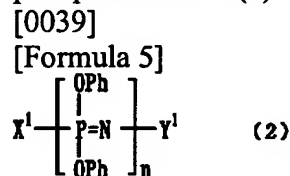
[0034] The loadings of a multilayer-structure polymer are 1 - 10 weight section to the aromatic series polycarbonate resin 100 weight section. If shock resistance is inadequate in a multilayer-structure polymer being under 1 weight section and 10 weight sections are exceeded, the fall of a mold-goods appearance will tend to take place. The loadings of a multilayer-structure polymer are 1 - 8 weight section preferably to the aromatic series polycarbonate resin 100 weight section, and are 2 - 5 weight section still more preferably.

[0035] As a phosphazene compound of the (d) component in this invention, into a molecule, it is a compound containing Lynn and nitrogen, and a well-known thing can be used widely conventionally.

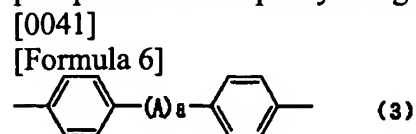
[0036] As a phosphazene compound, it is (1), for example. General formula (1)



[0038] The integer of 3-25 is shown by the inside m of [type. Ph shows a phenyl group.] The annular phenoxy phosphazene and (2) which are come out of and expressed General formula (2)



[0040] Radical-N=P (OPh)3 or radical-N=P(O) OPh is shown by the inside X1 of [type, and Y1 is a radical. - P(OPh) 4 or radical-P(O) (OPh)2 are shown. n shows the integer of 3-10000. Ph is the same as the above.] It is (3) to the chain-like phenoxy phosphazene and the list which are come out of and expressed. At least one sort of phosphazene compounds chosen from the group which consists of said annular phenoxy phosphazene and said chain-like phenoxy phosphazene are o-phenylene group, m-phenylene group, p-phenylene group, and a general formula (3).



[0042] -C(CH3)2-, -SO2-, -S-, or -O- is shown by the inside A of [type. a shows 0 or 1.] It is the compound over which at least one sort of bridge formation radicals chosen from the group which comes out and consists of a bis-phenylene group expressed come to construct a bridge. (i) This bridge formation radical intervenes between two oxygen atoms from which the phenyl group of a phosphazene compound was desorbed. (ii) The content rate of a phenyl group can mention the bridge formation phenoxy phosphazene compound which is 50 - 99.9%, and (iii) does

not have the free hydroxyl group in intramolecular on the basis of the total of the above-mentioned phosphazene compound (1) and/or all the phenyl groups in (2).

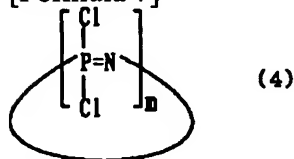
[0043] In this invention, "not having the free hydroxyl group in intramolecular" means that the free amount of hydroxyl groups is below limit of detection, when a quantum is carried out [page / of an analytical chemistry handbook / 353rd / (the 3rd edition of revision, edited by Japan Society for Analytical Chemistry, Maruzen Co., Ltd., 1981)] according to the acetylating method by the acetic anhydride and pyridine of a publication. Limit of detection is the limit of detection as a hydroxyl equivalent per 1g (bridge formation phenoxy phosphazene compound of this invention) of samples, and is more specifically below 1×10^{-6} hydroxyl equivalent / g here.

[0044] In addition, if the bridge formation phenoxy phosphazene compound of this invention is analyzed by the above-mentioned acetylating method, the amount of the hydroxyl group of the raw material phenol which remains will also be added, but since a raw material phenol can carry out a quantum with high performance chromatography, the quantum only of the free hydroxyl group in a bridge formation phenoxy phosphazene compound can be carried out.

[0045] As a manufacturing method of a bridge formation phenoxy phosphazene compound, it is a general formula (4), for example.

[0046]

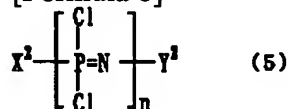
[Formula 7]



[0047] [-- the same as the above the inside m of a formula.] The annular dichlorophosphagen compound and general formula (5) which are come out of and expressed

[0048]

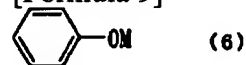
[Formula 8]



[0049] As for the inside X² of [type, radical-N=PCl₃ or radical-N=P(O) Cl is shown, and Y² shows radical-P(Cl)₄ or radical-P(O) Cl₂. n is the same as the above.] To at least one sort of dichlorophosphagen compounds chosen from the group which comes out and consists of a chain-like dichlorophosphagen compound expressed, it is a general formula (6).

[0050]

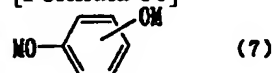
[Formula 9]



[0051] [-- alkali metal, such as a lithium, sodium, or a potassium, is shown by the inside M of a formula.] The alkali-metal phenolate come out of and expressed, and a general formula (7)

[0052]

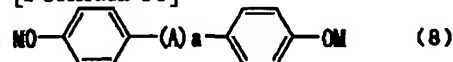
[Formula 10]



[0053] [-- the same as the above the inside M of a formula.] o- come out of and expressed, m-, p-permutation alkali-metal JIFENORATO, and a general formula (8)

[0054]

[Formula 11]



[0055] [-- the same as the above the inside A, a, and M of a formula.] The method of making mixture with at least one sort of JIFENORATO chosen from the group which comes out and consists of alkali-metal JIFENORATO expressed react (the first process), and making the above-mentioned alkali-metal phenolate react to the compound subsequently obtained further (the second process) is mentioned.

[0056] The general formula (4) and the dichlorophosphazene compound of (5) which are used as one of the raw materials by the above-mentioned manufacturing method are for example, H.R.Allcock work, "Phosphorus-Nitrogen Compounds", and Academic. Press, (1972), J.E.M ark, H.R.Allcock, R.West work, "Inorganic Polymers", Prentice-Hall International According to the approach of a publication (1992), it can manufacture to Inc. etc. That is, the dichlorophosphazene compound of the general formula (5) the dichlorophosphazene compound of the general formula (4) whose m is 3-25, and whose n are 3-25 can be manufactured by making an ammonium chloride and a phosphorus pentachloride (or an ammonium chloride, a phosphorus trichloride, and chlorine) react at about 120-130 degrees C, and, for example, carrying out demineralization oxidization in chlorobenzene or tetrachloroethane, first. These dichlorophosphazene compounds (dichlorophosphazene oligomer) are usually obtained as mixture. Moreover, annular dichlorophosphazene compounds and hexachlorocyclotriphosphazene, such as hexachlorocyclotriphosphazene, octachlorocyclotetraphosphazene, and deca KURORU cyclopentaphosphazene, are separable from the dichlorophosphazene oligomer mixture of the shape of annular [which was obtained by doing in this way], and a chain with distillation or recrystallization. Furthermore, n can manufacture the dichlorophosphazene compound expressed with the general formula (5) which is 25 to 10000 by heating and carrying out ring opening polymerization of these to 220-250 degrees C. It may dissociate and these dichlorophosphazene compounds may be respectively used independently, while the dichlorophosphazene of the shape of annular and a chain had been mixed.

[0057] As an alkali-metal phenolate expressed with a general formula (6), a well-known thing can be used widely conventionally, for example, a sodium phenolate, a potassium phenolate, a lithium phenolate, etc. can be mentioned. One sort can be independently used for these alkali-metal phenolate, or it can use two or more sorts together.

[0058] In alkali-metal JIFENORATO expressed with a general formula (7), two radical-O-M (M is the same as the above.) may be in alt.** meta or which physical relationship of Para. As an example of this alkali-metal JIFENORATO, alkali-metal salts, such as resorcinol, hydroquinone, and a catechol, can be mentioned, for example. Also in these, sodium salt and lithium salt are desirable. This alkali-metal JIFENORATO can use one sort independently, or can use two or more sorts together.

[0059] As alkali-metal JIFENORATO expressed with a general formula (8), alkali-metal salts, such as - oxy-diphenol, and - sulfonyl diphenol (bisphenol S), and 4 and 4'-isopropylidene diphenol (bisphenol A), 4, and 4 '4, 4'-thiodiphenol, 4, and 4 '4, 4'-diphenol, etc. can be mentioned, for example. Also in these, sodium salt and lithium salt are desirable. This alkali-metal JIFENORATO can use one sort independently, or can use two or more sorts together.

[0060] In this invention, alkali-metal JIFENORATO expressed with alkali-metal JIFENORATO and the general formula (8) which are expressed with a general formula (7) may be used independently, respectively, and these may be mixed and used.

[0061] In the first process of the manufacturing method of said bridge formation phosphazene, it is desirable to adjust the amount of an alkali-metal phenolate and the alkali-metal JIFENORATO used so that the chlorine atom in a dichlorophosphazene compound may not be altogether consumed by the reaction with an alkali-metal phenolate and alkali-metal JIFENORATO, namely, so that the chlorine atom in a dichlorophosphazene compound may remain in addition also by the reaction with an alkali-metal phenolate and alkali-metal JIFENORATO. Thereby, both-O-M radical (M is the same as the above.) of alkali-metal JIFENORATO combines with the Lynn atom of a dichlorophosphazene compound. What is necessary is just to usually make preferably about 0.05-0.9Eq of the amount of an alkali-metal phenolate and the alkali-metal JIFENORATO used into about 0.1-0.8Eq in the sum total of both phenolates on the basis of the chlorinity of a dichlorophosphazene compound in the first process.

[0062] In the second process of the manufacturing method of said bridge formation phosphazene, it is desirable to adjust the amount of the alkali-metal phenolate used so that the chlorine atom in the compound generated at the first process of the above may be altogether consumed by the reaction with an alkali-metal phenolate. What is necessary is just to usually make preferably about 1-1.5Eq of the amount of the alkali-metal phenolate used into about 1-1.2Eq on the basis of the chlorinity of a dichlorophosphazene compound in that case.

[0063] moreover, the operating rate (alkali-metal JIFENORATO / alkali-metal phenolate, mole ratio) of an alkali-metal

phenolate (total quantity used at the first process and the second process), and alkali-metal JIFENORATO -- usually -- about 1 / 2000 to 1/4 -- desirable -- 1 / 20 - 1/6 -- then, it is good.

[0064] the reaction of the first process and the second process -- each usual room temperature - it is preferably carried out to the bottom of the temperature of about 80-140 degrees C, and about 150 degrees C is usually completed preferably in about 3 - 7 hours for about 1 to 12 hours. Each reaction of the first process and the second process is usually performed in organic solvents, such as halogenated-aromatics hydrocarbons, such as aromatic hydrocarbon, such as benzene, toluene, and a xylene, mono-chlorobenzene, and dichlorobenzene.

[0065] The bridge formation phenoxy phosphazene compound manufactured by the above-mentioned reaction follows the usual isolation approaches, such as washing, filtration, and desiccation, and can be easily isolated and refined from a reaction mixture.

[0066] Thus, the obtained bridge formation phenoxy phosphazene compound usually has decomposition temperature in the range which is 250-350 degrees C.

[0067] Moreover, the content percentage of the phenyl group in this bridge formation phenoxy phosphazene compound is 50 - 99.9% on the basis of the total of all the phenyl groups in the annular phenoxy phosphazene compound of a general formula (1), and/or the chain-like phenoxy phosphazene compound of a general formula (2), and is usually 70 - 90% preferably.

[0068] in addition, when the end groups X1 and Y1 in a general formula (2) change with reaction conditions etc., and are the usual reaction conditions, for example, a mild reaction is performed by the system of non-water X1 is radical-N=P (OPh)₃ and Y1 is a radical. - by reaction condition to which it becomes the structure of P(OPh)₄, and moisture or an alkali-metal hydroxide exists in the system of reaction Or when it reacts by severe reaction condition which a transition reaction produces, X1 is radical-N=P (OPh)₃ and Y1 is a radical. - It will be in the condition that radical-N=P (O) OPh is intermingled by X1 and the thing of the structure of radical-P(O) (OPh)₂ is intermingled by Y1 other than the structure of P(OPh)₄.

[0069] (d) The phenoxy phosphazene compound of the above-mentioned various kinds as a phosphazene compound of a component, Namely, the annular phosphazene compound expressed with a general formula (1), the chain-like phosphazene compound expressed with a general formula (2), A part of phenyl group in annular [which is expressed with said general formula (1) and (2)] and a chain-like phosphazene compound And o-phenyl group, The bridge formation phenoxy phosphazene compound replaced by the bridge formation radical expressed with m-phenyl group, p-phenyl group, and said general formula (3) can be used preferably.

[0070] Annular [of these various kinds], the shape of a chain, and a bridge formation phenoxy phosphazene compound do not contain a halogen. Therefore, in case the resin constituent of this invention which used this decomposes or burns, it does not generate harmful gas or smoke to a living thing, such as hydrogen halide. Moreover, the corrosion of metal mold, degradation of resin, and coloring are not caused at the time of resin shaping. Moreover, the various aforementioned phenoxy phosphazene compounds preferably used for this invention do not reduce the molding temperature of resin, and its volatility is low and they do not have blocking or that ooze out (JUSHINGU) and problems, such as dripping at the time of combustion, arise at the time of kneading.

[0071] Moreover, since this thing is the bridge formation phenoxy phosphazene compound which does not have the one end hydroxy group of a dihydroxy compound substantially when using a bridge formation phenoxy phosphazene compound as a (d) component, molecular weight of resin is not reduced and the properties of resin original, such as mechanical properties, such as shock resistance, thermal resistance, and fabrication nature, are not reduced.

[0072] In this invention, the loadings of (d) phosphazene compound are 0.5 - 40 weight section to the aromatic series polycarbonate resin 100 weight section. If fire retardancy is inadequate in the addition of a phosphazene compound being under the 0.5 weight section and 40 weight sections are exceeded, mechanical physical properties will tend to fall. the loadings of a phosphazene compound -- the aromatic series polycarbonate resin 100 weight section -- receiving -- desirable -- 1 - 30 weight section -- it is 2 - 25 weight section still more preferably.

[0073] The inclination which the polytetrafluoroethylene which has fibril organization potency, for example is mentioned as poly fluoro ethylene used as a (e) component in this invention, and distributes easily in a polymer, and combines polymers, and makes a fibrous ingredient is shown. The polytetrafluoroethylene which has fibril organization potency is classified into Type 3 according to ASTM specification.

[0074] As polytetrafluoroethylene which has fibril organization potency, it is marketed from Dupont-Mitsui Fluorochemicals, Inc., and is marketed, for example as Pori Flon from Daikin Industries, LTD. as Teflon 6J or Teflon

30J.

[0075] The loadings of poly fluoro ethylene are 0.01 - 5 weight section to the aromatic series polycarbonate resin 100 weight section. If fire retardancy is inadequate in poly fluoro ethylene being under the 0.01 weight section and 5 weight sections are exceeded, the fall of a mold-goods appearance will tend to take place. The loadings of poly fluoro ethylene are 0.02 - 4 weight section preferably to the aromatic series polycarbonate resin 100 weight section, and are 0.03 - 3 weight section still more preferably.

[0076] As the manufacture approach of the polycarbonate resin constituent of this invention, the approach of blending aromatic series vinyl / vinylcyanide system copolymer, a multilayer-structure polymer, and polytetrafluoroethylene after kneading and with this, and carrying out melting kneading at them etc. is beforehand mentioned in the approach and aromatic series polycarbonate resin which especially a limit does not have, for example, carry out package melting kneading of aromatic series polycarbonate resin, aromatic series vinyl / vinylcyanide system copolymer, a multilayer-structure polymer, a phosphazene compound, and the polytetrafluoroethylene, and a phosphazene compound.

[0077] The addition combination of the whiskers, such as reinforcement, such as additives, such as stabilizers, such as an ultraviolet ray absorbent and an antioxidant, a pigment, a color, lubricant, other flame retarders, a fire-resistant assistant, a release agent, and a sliding nature amelioration agent, a glass fiber, a glass flake, and a carbon fiber, and potassium titanate, boric-acid aluminum, can be carried out at the polycarbonate resin constituent of this invention if needed. There is also a mode of the resin constituent with which the ultraviolet ray absorbent is not blended in the polycarbonate resin constituent of this invention.

[0078] In the fire-resistant polycarbonate resin constituent of this invention, it is the range which does not spoil the property and thermoplastics, such as polyester resin like polybutylene terephthalate and polyethylene terephthalate, polyamide resin, and polyolefin resin, can be blended. The loadings of such thermoplastics are 30 or less % of the weight more preferably 40 or less % of the weight to the resinous principle whole quantity in the resin constituent of this invention.

[0079] The fire-resistant polycarbonate resin constituent of this invention is a fire-resistant polycarbonate resin constituent which comes to blend a non-halogen phosphazene compound, and since there are few of it being a non-halogen, respectively or halogen contents, the component blended in the resin constituent of this invention is an ingredient desirable when abolishing the corrosion problem of a making machine metallurgy mold.

[0080] The fire-resistant polycarbonate resin constituent of this invention can be used as a fire-resistant polycarbonate resin Plastic solid by applying the usual shaping means.

[0081]

[Effect of the Invention] The fire-resistant polycarbonate resin constituent of this invention is excellent in fire retardancy, shock resistance, and a fluidity, and excellent in the thermal resistance and the moisture resistance which can maintain high physical properties an elevated temperature and under highly humid.

[0082] The polycarbonate resin constituent of this invention is excellent in fire retardancy, excellent in the mechanical strength under heat and high humidity, and useful as the electrical and electric equipment, the large-sized mold goods in the precision instrument field, or light-gage mold goods.

[0083]

[Example] Hereafter, although an example explains this invention to a detail further, this invention is not limited to the following examples, unless the summary is exceeded.

[0084] In a following example and the following example of a comparison, the raw material of a publication was used next.

PC resin -1 : (1) Polly 4, 4-isopropylidene diphenyl carbonate, The you pyrone S-3000, the product made from Mitsubishi Engineering plastics, Viscosity-average-molecular-weight 21000(2) PC resin -2 : Polly 4, 4-isopropylidene diphenyl carbonate, The you pyrone S-3000, the product made from Mitsubishi Engineering plastics, Viscosity-average-molecular-weight 21000 (3) ABS plastics : CBT-698, ABS-plastics (4) AS-resin:SAN-C made from Techno Polymer, A techno polymer Make AS resin (5) Multilayer-structure polymer-1:EXL2603 Multilayer-structure polymer (6) multilayer-structure polymer -2:E-901 of the butadiene core by Kureha Chemical Industry Co., Ltd. / acrylic shell Multilayer-structure polymer (7) multilayer-structure polymer-3:SRK-200 of a butadiene [by Mitsubishi Rayon Co., Ltd.] d styrene core / acrylic shell Silicone content acrylic-ester system multilayer-structure polymer (8) flame retarder by Mitsubishi Rayon Co., Ltd.: Condensed-phosphoric-acid ester PX-200, the product made from Daihachi Chemistry. (9) Polytetrafluoroethylene (PRFE) : Pori Flon F-201L, Daikin, LTD. make.

[0085] In addition, physical-properties evaluation of a test piece was performed next like a publication.

(10) Flammability : the test piece of UL of 1.6mm thickness performed the perpendicular combustion test, and it evaluated.

(11) Impact strength : the 3.2mm test piece for Izod impact test was fabricated, and it evaluated by cutting the notch of 0.25R after that.

(12) Fluidity : it is 240 degrees C with a quantity-sized type flow tester. The conditions of 160 kg/cm² estimated by the 1mm orifice.

(13) Weld reinforcement : the piece of anti-tension which has the weld section was used, **** fracture strength was measured, and the **** fracture strength in the piece of anti-tension without the weld section was compared.

(14) Moisture resistance : 60 degrees C After holding the piece of anti-tension without the weld section for 200 hours, the tension test was performed, and **** elongation in humidity resistance test order was compared with the bottom of 90% of condition (unit %).

[0086] The synthetic example 1 (composition of the phenoxy phosphazene compound which has the structure of cross linkage by PARAFENIREN)

The toluene solution of a sodium phenolate was adjusted by carrying out the heating reflux of a 1.1 mols (103.5g) phenol, a 1.1 mols (44.0g) sodium hydroxide, 50g of water, and the toluene 500ml mixture, and removing only water besides a system.

[0087] The toluene solution of the lithium salt of hydroquinone and a phenol was prepared by putting 0.15 mols (16.5g) hydroquinone, a 1.0 mols (94.1g) phenol, a 1.3 mols (31.1g) lithium hydroxide, 52g of water, and toluene 600ml mixture into a 2l. 4 opening flask, carrying out heating reflux in parallel to said reaction, and removing only water besides a system. After 580g of 20% chlorobenzene solutions which contain a dichlorophosphazene oligomer (62% [of trimers], 12% [of tetramers], pentamer and 11% [of hexamers], 3% [of 7 ****], and more than octamer 12% of mixture) 1.0 unit mol (115.9g) in this toluene solution was dropped below 30 degrees C under stirring, the stirring reaction was carried out at 110 degrees C for 3 hours. Next, after adding under stirring the toluene solution of a sodium phenolate adjusted previously, the reaction was continued at 110 degrees C for 4 hours.

[0088] After 1.0l. of sodium-hydroxide water solutions washed the reaction mixture 3 times 3% after reaction termination, next 1.0l. of water washed 3 times, the organic layer was condensed under reduced pressure. The heating vacuum drying of the obtained product was carried out by 80 degrees C and 3 mmHg or less for 11 hours, and 211g fine yellow powder was obtained.

[0089] The hydrolysis chlorine of the bridge formation phenoxy phosphazene compound obtained above was 0.04%, weight average molecular weight (Mw) was 1080 in polystyrene conversion (based on GPC analysis), and the presentation of the last object was [N=P(-O-p-Ph-O-) 0.15 (-O-Ph) 1.7] from the CHN elemental-analysis value at the Lynn content list.

[0090] Weight average molecular weight (Mw) was 1100 in polystyrene conversion (based on GPC analysis), TG/DTA analysis did not show the clear melting point, but 306 degrees C and 5% weight reduction temperature of decomposition initiation temperature were 311 degrees C.

[0091] Moreover, as a result of performing the quantum of a residual hydroxy group by the acetylating method, it was below limit of detection (considering as the hydroxy equivalent per sample 1g : 1x10⁻⁶Eq/(g) or less).

[0092] The synthetic example 2 (composition of the phenoxy phosphazene compound which has the structure of cross linkage by 2 and 2-screw (p-oxy-phenyl) isopropylidene radical)

0.7 mol [of phenols] (65.9g) and toluene 500ml was put into the 1l. 4 opening flask, and under stirring, keeping internal solution temperature at 25 degrees C, the metallic-sodium 0.65 gram atom (14.9g) was cut out finely, and was thrown in. Stirring was continued for 8 hours until metallic sodium disappeared completely at 77-113 degrees C after injection termination.

[0093] In parallel to said reaction, 0.25 mol [of bisphenol A] (57.1g), 1.1 mol [of phenols] (103.5g), and tetrahydrofuran (THF) 800ml was put into the 3l. 4 opening flask, and under stirring, keeping internal solution temperature at 25 degrees C, the metal lithium 1.6 gram atom (11.1g) was cut out finely, and was thrown in. Stirring was continued for 8 hours after injection termination until the metal lithium disappeared completely at 61-68 degrees C. Under stirring of 1.0 mols (37%, 313g of chlorobenzene solutions, a presentation: concentration : 75% of trimers, 17% of tetramers, a pentamer and 6% of hexamers, 1% of 7 ****, and more than an octamer 1% of mixture) (115.9g) of dichlorophosphazene oligomers in this slurry solution, after being dropped over 1 hour, keeping internal solution

temperature at 20 degrees C or less, it reacted at 80 degrees C for 2 hours. Subsequently, under stirring, after adding the sodium-phenolate solution adjusted separately over 1 hour, keeping internal solution temperature at 20 degrees C, it reacted at 80 degrees C for 5 hours.

[0094] After reaction termination, the reaction mixture was condensed and 1l. of toluene was newly added except for THF. It is 2%NaOH about this toluene solution. After it washed 3 times by 1l., next 1l. of water washed 3 times, the organic layer was condensed under reduced pressure. The heating vacuum drying of the obtained product was carried out by 80 degrees C and 3 mmHg or less for 11 hours, and 229g white powder was obtained.

[0095] The presentation of the last object of the hydrolysis chlorine of the bridge formation phenoxy phosphazene compound obtained above was $[N=P(-O-Ph-C(CH_3)_2-Ph-O-) 0.25 (-O-Ph) 1.50]$ from the CHN elemental-analysis value 0.07% at the Lynn content list.

[0096] Weight average molecular weight (Mw) was 1130 in polystyrene conversion (based on GPC analysis), TG/DTA analysis did not show the clear melting point, but 308 degrees C and 5% weight reduction temperature of decomposition initiation temperature were 313 degrees C.

[0097] Moreover, as a result of performing the quantum of residual hydroxyl by the acetylating method, it was below limit of detection (considering as the hydroxyl equivalent per sample $1g : 1 \times 10^{-6} Eq/(g)$ or less).

[0098] The synthetic example 3 (composition of the phenoxy phosphazene which has the structure of cross linkage by 4 and 4-sulfonyl diphenylene (bisphenol-S residue))

0.4 mols (37.6g) of phenols, THF 500ml was put into the 1l. 4 opening flask, and under stirring, keeping internal solution temperature at 25 degrees C, the metallic-sodium 0.45 gram atom (9.2g) was cut out finely, and was thrown in. Stirring was continued for 5 hours until metallic sodium disappeared completely at 65-72 degrees C after injection termination.

[0099] It is concurrent with said reaction and they are 1.70 mols (160.0g) of phenols, and a bisphenol with a 1l. 4 opening flask. - S 0.05 mols (12.5g) were dissolved in tetrahydrofuran (THF) 500ml, the metallic-sodium 1.8 gram atom (41.4g) was thrown in below 25 degrees C, stirring was continued at a temperature up and 61 degrees C - 68 degrees C to 61 degrees C over 1 hour for 6 hours after injection termination, and the sodium-phenolate mixed solution was adjusted. The stirring reaction of this solution was carried out at 71-73 degrees C after dropping under cooling / stirring of 25 degrees C or less for 5 hours at 580g of 20% chlorobenzene solutions containing a dichlorophosphazene oligomer (62% [of trimers], 12% [of tetramers], pentamer and 11% [of hexamers], 3% [of 7 ****], and more than octamer 12% of mixture) 1.0 unit mol (115.9g).

[0100] Next, after the sodium-phenolate mixed solution prepared previously was dropped, the reaction was continued at 71-73 degrees C for 3 hours.

[0101] After condensing the reaction mixture after reaction termination and remelting to chlorobenzene 500ml, 3 times, 5% sulfuric acid treatment, 5% sodium bicarbonate backwashing by water, and three rinsings were performed, concentration hardening by drying of the 5%NaOH backwashing by water was carried out, and 218g of wax-like objects of light yellow was obtained.

[0102] The hydrolysis chlorine of the bridge formation phenoxy phosphazene compound obtained above is 0.01% or less, and the presentation of this object was mostly determined as $[N=P(-O-Ph-SO_2-Ph-O-) 0.05 (-O-Ph) 1.90]$ from the CHN elemental-analysis value at the phosphorus content list.

[0103] Weight average molecular weight (Mw) was 1080 in polystyrene conversion, and the melting out temperature (Tm) by TG/DTA analysis was [320 degrees C and 5% weight reduction temperature of 103 degrees C and decomposition initiation temperature] 334 degrees C.

[0104] Moreover, as a result of performing the quantum of a residual hydroxy group by the acetylating method, it was below limit of detection (considering as the hydroxy equivalent per sample $1g : 1 \times 10^{-6} Eq/(g)$ or less).

[0105] The phosphazene compound of the ABS plastics of 25 weight sections, the multilayer-structure polymer -1 of 7 weight sections, and the synthetic example 1 of 15 weight sections and the poly fluoro ethylene of the 0.4 weight section were blended with PC resin of the example 1100 weight section, and it pelletized 240 degrees C of cylinder temperatures with the 30mm twin screw extruder after 20-minute mixing with the tumbler, and the piece of a combustion test of 1.6mm thickness was fabricated with the injection molding machine, and it estimated fire retardancy.

[0106] Furthermore, the test piece for Izod impact test, the piece of anti-tension which has the weld section, and the piece of anti-tension which does not have the weld section were fabricated, and after that, the test piece for Izod impact

test cut the notch of 0.25R by the notching machine, and evaluated by 240 degrees C of cylinder temperatures with the Izod impact tester. After carrying out conditioning of the piece of anti-tension which does not have the weld section on condition that (14), it performed the tension test. An evaluation result is shown in Table 1.

[0107] In example 2 example 1, except changing the multilayer-structure polymer -1 into the multilayer-structure polymer -2, it pelletized by the same approach as an example 1, and evaluated similarly. A result is shown in Table 1.

[0108] In example 3 example 1, except changing the ABS plastics of 25 weight sections into the ABS plastics of 17 weight sections, and the AS resin of 8 weight sections, it pelletized by the same approach as an example 1, and evaluated similarly. A result is shown in Table 1.

[0109] In example 4 example 1, except changing the multilayer-structure polymer -1 into the multilayer-structure polymer -3, it pelletized by the same approach as an example 1, and evaluated similarly. A result is shown in Table 1.

[0110] In example 5 example 1, except changing the phosphazene compound of the synthetic example 1 into the phosphazene compound of the synthetic example 2, it pelletized by the same approach as an example 1, and evaluated similarly. A result is shown in Table 1.

[0111] In example 6 example 1, except changing the phosphazene compound of the synthetic example 1 into the phosphazene compound of the synthetic example 3, it pelletized by the same approach as an example 1, and evaluated similarly. A result is shown in Table 1.

[0112] In example of comparison 1 example 1, it evaluated by being the same approach as an example 1 except not adding the multilayer-structure polymer -1. A result is shown in Table 1.

[0113] In the example 1 of example of comparison 2 comparison, PC resin -1 was changed into PC resin -2, and it evaluated by being the same approach as an example 1 except not adding the multilayer-structure polymer -1. A result is shown in Table 1.

[0114] In example of comparison 3 example 1, except changing the phosphazene compound of the synthetic example -1 into phosphoric ester, it pelletized by the same approach as an example 1, and evaluated similarly. A result is shown in Table 1.

[0115]

[Table 1]

	実施例 1	実施例 2	実施例 3	実施例 4	実施例 5	実施例 6	比較例 1	比較例 2	比較例 3
PC樹脂-1	100	100	100	100	100	100	100	—	100
PC樹脂-2	—	—	—	—	—	—	—	100	—
ABS樹脂	25	25	17	25	25	25	25	25	25
AS樹脂	—	—	8	—	—	—	—	—	—
多層構造重合体-1	7	—	7	—	7	7	—	—	7
多層構造重合体-2	—	7	—	—	—	—	—	—	—
多層構造重合体-3	—	—	—	7	—	—	—	—	—
合成例 1	15	15	15	15	—	—	15	15	—
合成例 2	—	—	—	—	15	—	—	—	—
合成例 3	—	—	—	—	—	15	—	—	—
燐酸エステル	—	—	—	—	—	—	—	—	15
PTFE	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4
流動性 ml/sec	24	24	28	25	24	24	26	13	22
アイゾット衝撃強さ	699	637	614	642	680	673	137	643	550
燃焼性 1.6mm	V-0	V-0	V-0	V-0	V-0	V-0	V-0	V-0	V-0
ウェルド強さ 無	63	61	60	60	63	61	63	63	61
ウェルド強さ 有	49	48	48	45	50	48	38	37	45
耐湿性 0時間	132	130	128	134	133	127	102	123	118
200時間	127	125	125	130	130	125	87	110	30

[0116] The following things are clear from the result of Table 1.

(1) In the example 1 of a comparison, if the ingredient which lowered the molecular weight of PC resin is used in order to secure a fluidity, impact strength will fall sharply. A fluid fall will become large, if the molecular weight of PC resin is raised in order to secure impact nature like the example 2 of a comparison on the other hand. Even if it lowered the molecular weight of PC resin from the example 1 in the example 4 to it, by adding a multilayer-structure polymer, impact strength was also held and the reinforcement of the weld section has also been improved further.

- (2) By damp-proof comparison, it has been sharply improved by using a phosphazene compound as compared with the constituent (example 3 of a comparison) which used conventional phosphoric ester.
- (3) These properties were maintained also for the phosphazene compound into which the structure of phosphazene was changed (an example 5 and example 6).
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[Translation done.]